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Research note

Calibration curves for on-line leakage detection using radiotracer injection method

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Abstract
One of the most important requirements for gas and fluid industrial pipelines is the on-line leakage detection and investigation. In this paper, detection of leak and determination of its amount using radioactive tracer injection method has been simulated by Monte Carlo MCNP code. The detector array included two NaI (Tl) detectors which were located before and after the considered position, measures emitted gamma from radioactive tracer. After calibration of radiation detectors, the amount of leakage can be calculated based on the count difference of areas under the radiation detectors. Also, the factors affecting the detectable amount of leakage such as the material, thickness, diameter of pipe, types of fluid, radiation detector crystal dimension, the activity of tracer and its type (^{24}Na, ^{82}Br, ^{131}I, ^{99m}Tc, ^{113m}In) have been investigated. According to the results, for example, leakage more than 0.007% in volume of the inlet fluid for iron pipe with outer diameter 4 inch and thickness of 0.5 cm, petrol as fluid inside pipe, 3×3 inch detector and ^{24}Na with activity of 100 mCi as detector can be detected by the presented method.

Keywords: leakage, radiotracer, MCNPX 2.7e, NaI(Tl), pipeline

1. Introduction

Leaking of petroleum or industrial flow in pipeline should be avoided because of economical aspect as well as protection of the environment against the petroleum flow. One of the fast and reliable methods of online investigation of pipelines is the radiotracer (gamma emitter radioisotope) injection method into the pipeline, which is rapidly mixed with liquid flow and proceeds. If a small amount of radiotracer gets into the environment, the activity of flow will be reduced. The measurement of this reduction is the key for leakage detection.

Some methods such as magnetic and acoustic methods can be applied *on* or *off-line* with the most common methods used being steady state equation, hydrostatic-testing, negative pressure wave, statistical analysis model, the traveling wave principle, frequency analysis, and inverse analysis [1]. There are several methods to find leaks but most of them (negative pressure wave, acoustic), limited space, affected by ambient noise around, are not suitable for PVC pipeline, finding the approximate or wrong location of the leak in PVC pipelines. Standing Wave Difference Method for Leak Detection in Pipeline Systems [1] finds the approximate location of the leak. Water leaks in supply pipes can be detected by continuous wave sensor operating at 2.45 GHz [2]. In this method, pipe existence in the deep down or wet soil is reduced by the accuracy of leak measurement.

The radiotracer injection method is also fast, online, precise and independent from some environmental factors such as pressure, temperature, fluid movement which are invisible and hard to control during measurement [3, 4].

According to the fluid velocity as well as radiotracer activity, online measurement can be performed for different lengths of pipeline. In several projects, ^{82}Br was used to find a leak in the high-pressure heat exchangers. In these projects, by placing detectors in various locations, the profile of response time of detectors as well as the position of leak in the heat exchanger was found [5-7]. Radiotracer of ^{41}Ar with different activities were used for leak detection in different parts of the heat exchanger [6, 8]. IAEA used 50 mCi of ^{131}I as a tracer to find leak in underground pipelines suspected for leaking. In that experimental work, 7 NaI(Tl) detectors with dimension of 2×2 inch were used [6].

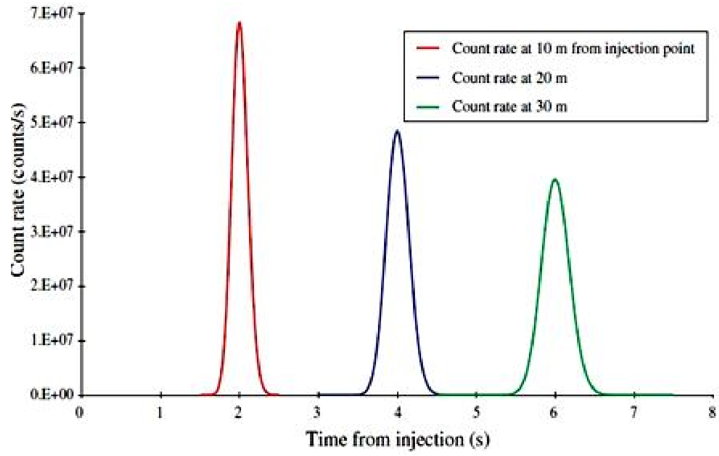

Figure 1. Distribution of tracer in different distances after the injection site.

Table 1. Radiotracers commonly used for leak detection in heat exchangers [9].

Radioisotope	Half-life	Gamma Energy, MeV (abundance %)	Chemical Form	Tracing Phase
^{24}Na	15 h	1.37 (100) 2.75 (100)	Sodium carbonate	Aqueous
^{82}Br	36 h	0.55 (70) 1.32 (27)	Ammonium bromide, Methylbromide, Dibromobenzene	Aqueous Gases Organic
^{131}I	8.04 d	0.36 (80) 0.64 (9)	Potassium or sodium iodide, Iodobenzene, Hippuran	Aqueous Organic
^{99m}Tc	6 h	0.14 (90)	Pertechnetate	Aqueous
^{113m}In	100 min	0.392 (65)	EDTA complex	Aqueous

In this paper, by placing two detectors before and after the considered length of pipe, gamma spectra are in detector array in order to monitor the radioactive concentration. For the determined duration, the net area under recognized peak is calculated and is recorded vs. time. These profiles of two detectors are compared. If there is no activity loss, the net areas under curves are equal and NAUC[1] of the gamma emitter are the same (Figure 1) [9].

In this paper, in addition to proposing the system for detection of leak, the amount of leakage per volume of inlet fluid has been investigated and the effect of some factors such as density of fluid, thickness of pipeline, type of radiotracer as well as its activity have been studied to determine the minimum detectable leakage in various conditions. Finally, calibration curves for various conditions have been presented that can be used for leak detection and can be generated according to the other working conditions.

2. Material and methods

As mentioned above, the radiotracer can be selected according to the working conditions. Gamma emitter radioisotopes such as ^{99m}Tc, ^{113m}In, ^{131}I, ^{82}Br, ^{24}Na have been considered in this study.

The selected radiotracers (Table 1) are gamma emitters with sufficient energy to penetrate as well as appropriate half-lives (not long half-life to be wiped out easily after completing the measurement and not short half-life to need the correction in count due to decay during measurement). Beta and alpha, due to their very short range, and neutron, because of physical health aspects, are not usually used as a radiotracer in these investigations. This method is based on the measurement of gamma rays; so, according to the economic reasons and easier performance, NaI(Tl) detectors have been selected. For efficient measurements, detector with various diameters of 1 to 8 inches as well as a height of 3 inch has been tested. figure 2 shows the schematic view of measurement configuration.

Gamma emission of radioactive fluid (fluid mixing with gamma radiotracer) that is passing in front of detectors has been considered as a criterion for measurement. Therefore, measurement (counting) before the first appearance of tracer in the detector is avoided by using cylindrical lead shield in the lateral surface of detectors (Figure 1).

According to the half value layer (HVL) of lead and largest photon energy in this investigation related to ^{24}Na (2.75 MeV), sufficient thickness of lead has been calculated (for 3 MeV (linear attenuation coefficient) μ =0.47 1/cm [10], so HVL=Ln2/μ =1.44 cm, 4*HVL =5.5 cm which means that only 1% of photons with this energy may be appeared in the detector). As an independent problem, in the worst case, if the radioactive fluid is in the pipe, the contribution of the gamma from other parts of pipe (not in front of the detector) in the count rate will be less than 1%. So, 5.5 cm lead shield has been considered surrounding each of detectors.

Photons are recorded during the determined time in

1. net area under curve

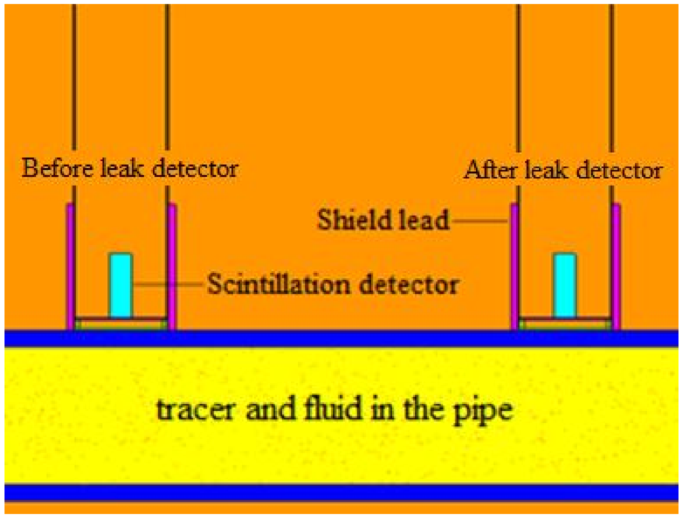

Figure 2. Schematic view of leakage measurement configuration in the pipe.

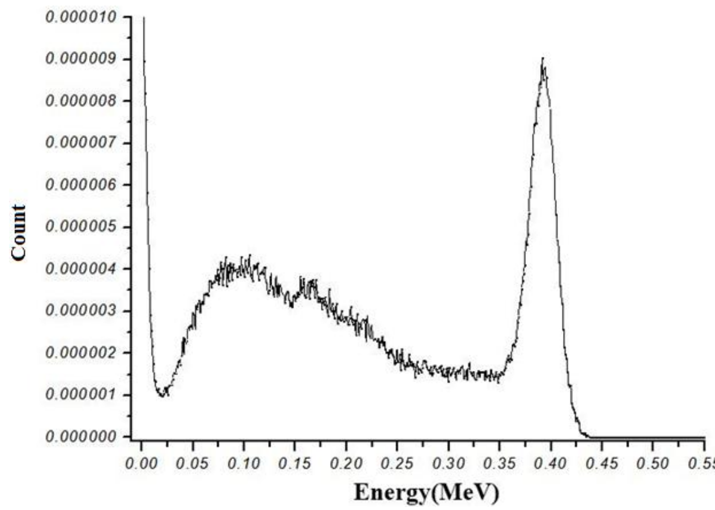

Figure 3. Spectrum of ^{113m}In in NaI(Tl) (PE pipe with thickness of 4 cm, outer diameter of 4 inch, petrol a fluid), vertical axis with arbitrary unit.

the detector's buffer as a count rate, so time profile of count for each detector can be achieved in terms of the movement of the tracer.

It should be mentioned that saturation of detector should be avoided, so the correct measurement can be obtained by selection of an appropriate activity for radiotracer. Figure 3 shows NaI(Tl) spectrum from ^{113m}In as a sample (PE pipe with thickness of 4 cm, outer diameter of 4 inch, petrol as a fluid) .

NAUC of count rate vs. time (due to the determined fluid velocity) can be converted to the total count of detector when the tracer completely passes in front of it. The existence of leakage causes the reduction in amount of radiotracer and consequently the reduction in the amount of activity. When the tracer reaches to the second detector, which is located after the leakage, the smaller NAUC compared to the NAUC measured by first detector indicates the presence of a leak.

There is a direct correlation between the reduced amount of radiotracer and the difference of NAUC in two detectors. By assuming the leakage positioned between two detectors, the amount of leakage can be measured independent of its location. All explained procedures have been simulated using MCNPX 2.7e (ENDF/B-VI Release 8 Photoatomic Data were used for photon and electron transport). It should be mentioned that relative error of all simulation results are less than 0.3%.

3. Results

Figure 4 shows the count rate profile vs. time for different amounts of leakage. More leakages cause less NAUC of count rate vs. time. As shown in figure 5, different amounts of leakage are investigated. Leakages more than 0.033% of the fluid volume can be determined with confidence level of 99.7% (3σ, equal to error of 0.3%). In fact, for the fluid flow of 10,000 liters per hour, leakage of 3.3 liter per hour can be measured and distinguished by the 2×3 inch detector using 10 mCi of ^{99m}Tc.

Some parameters such as density of fluid (with various densities; petrol, gasoline, white oil, water), thickness of pipeline, type of radiotracer as well as its

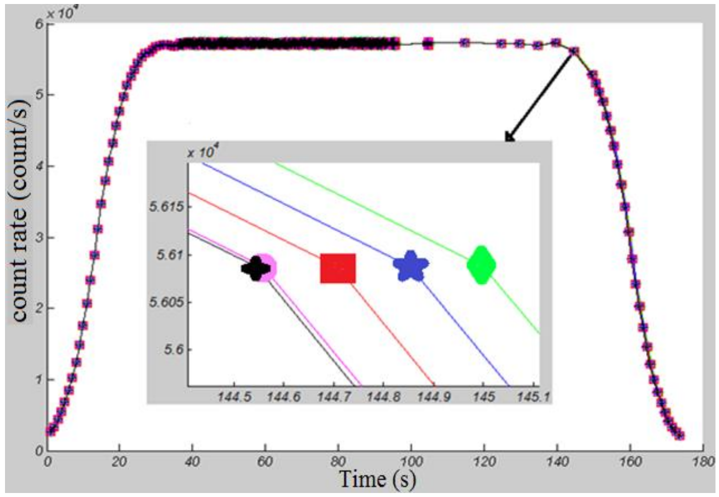

Figure 4. Ccurve count rate vs. time in iron pipe (wall thickness of 0.5, diameter of 4 inch and radiotracer of ^{99m}Tc with activity 10 mCi), ♦ : no leakage , ★: 0.1% , ■ : 0.2% , ● : 0.3% , ✚ : 0.31% Leakage in volume.

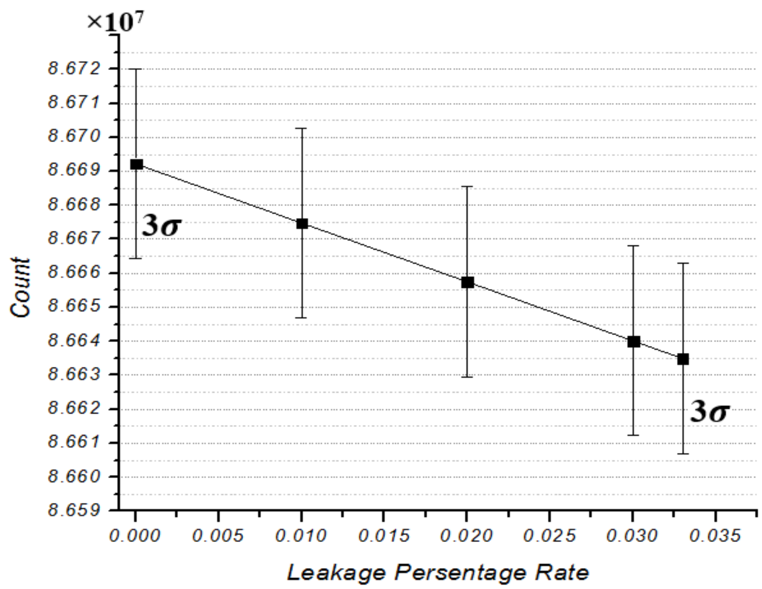

Figure 5. Determination of minimum detectable leakage using ^{99m}Tc with activity 10 mCi.

activity have been studied to determine the minimum detectable leakage.

3.1. Effect of fluid density in the leakage detection and determination of its amount (10mCi of ^{99m}Tc)

By increasing the density of fluid, photons received by the detector will be reduced due to the self-absorption in the fluid. In a similar measurement conditions, more count rate in detector results smaller leaks detection, so better distinguishing will be obtained in lower density fluid. Count rate in detector is dependent on the received photons to the detector, so less fluid density means more gammas transmission, which results better leakage diagnosis.

In addition, dimension of detectors (height and diameter in cylindrical geometry) have been investigated due to their effects on counting efficiency and consequently in measuring the leakage (Figure 6). It can be seen that by increasing the diameter that induces larger efficiency, the number of photons detected in detector will be increased, so less leakages can be detected well.

For detectors with dimensions of 1×3 inch, leakages more than 0.053% and 0.054% of inlet fluid volume can be distinguished for petrol (0.7 g/cm^3) and white oil (0.9 g/ cm^3), respectively.

For detectors with dimensions of 3×3 inch, leakages more than 0.025% of inlet fluid volume can be distinguished for petrol (0.7 g/cm^3) and white oil (0.9 g/ cm^3) (Figure 6). Less leakage can be measured by increasing the diameter of detector.

Determination the amount of leakage depends on the pipe material. According to figure 7, better leakage determination can be distinguished in polyethylene pipe in comparison to iron pipe.

For detectors with dimensions of 3×3 inch and polyethylene pipe, leakages more than 0.015% of input volume can be distinguished for petrol (0.7 g/cm^3) and

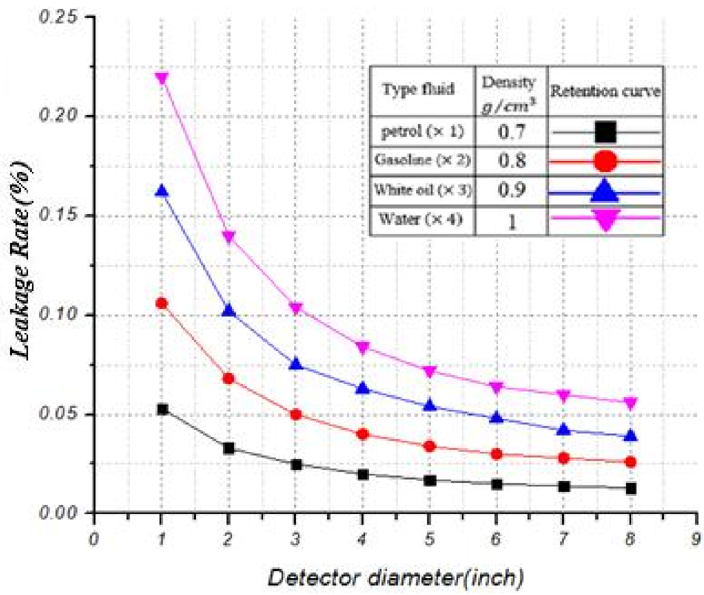

Figure 6. The effect of fluid density in iron pipe on detectable leakage using ^{99m}Tc with activity of 10 mCi.

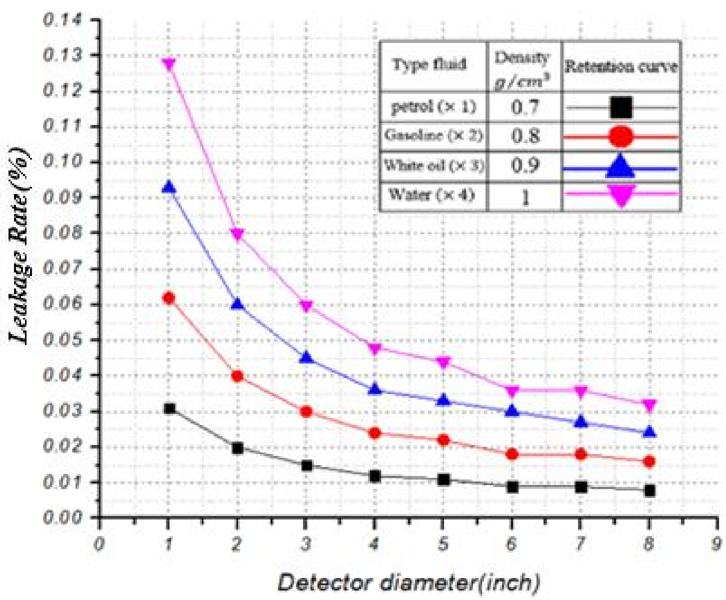

Figure 7. The effect of fluid density in polyethylene pipe on detectable leakage using ^{99m}Tc with activity of 10 mCi.

white oil (0.9 g/ cm^3).

3.2. Effect of thickness of pipe in the leakage detection and determination of its amount (10mCi of ^{99m}Tc)

By increasing the wall thickness of pipe, the number of photons reaching to the detector will be decreased. HVL of iron in energy of 0.14 MeV is 0.493 cm [10]. The effect of iron pipe with outer diameter of 4 inch and petrol as a fluid is shown in figure 8.

According to the results shown in figure 8, for the pipe with wall thickness of 4 cm (outer diameter of 4 inches, detector of 1×3 inch, petrol as a fluid and ^{99m}Tc as a tracer with activity 10 mCi) leakages more than 1.091% of inlet fluid volume can be measured. By using the detectors with larger diameter, the less leakage (better measurement) can also be measured. According to. figure 9, increasing the thickness of polyethylene pipe has less effect on the amount of detectable leakage because of its HVL (in energy of 0.14 MeV is 4.902 cm). For the polyethylene pipe with wall thickness of 1 and 4 cm (outer diameter of 4 inches, detectors of 3×3 inch, petrol as a fluid and ^{99m}Tc as a tracer with activity 10 mCi) leakages more than 0.016 % and 0.018% of inlet fluid volume can be measured, respectively.

3.3. Effect of diameter of pipeline in the leakage detection and determination of its amount (10mCi of ^{99m}Tc)

Increasing the diameter of the pipe increases the absorption of photons emitted by tracer in the fluid (self-shielding effect of fluid), so fewer photons can reach to

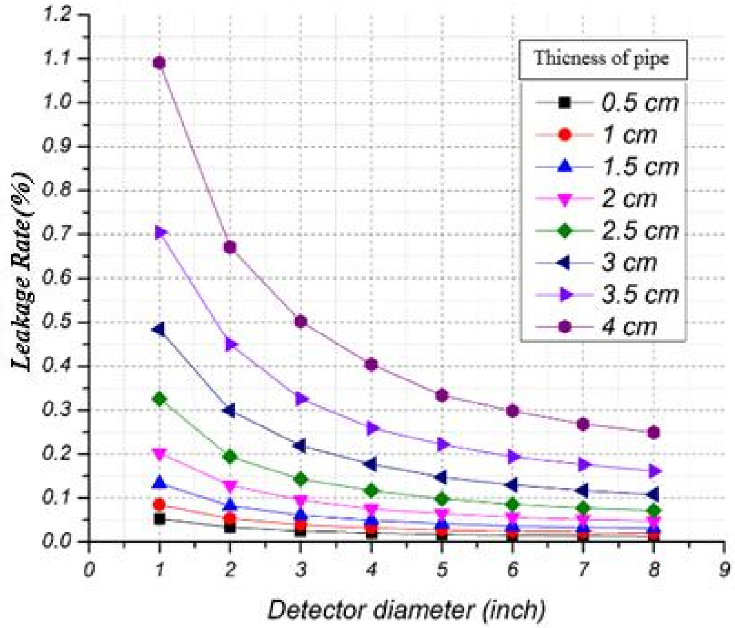

Figure 8. The effect of iron thickness (pipe diameter 4 inch) on the detectable leakage using ^{99m}Tc with activity of 10 mCi.

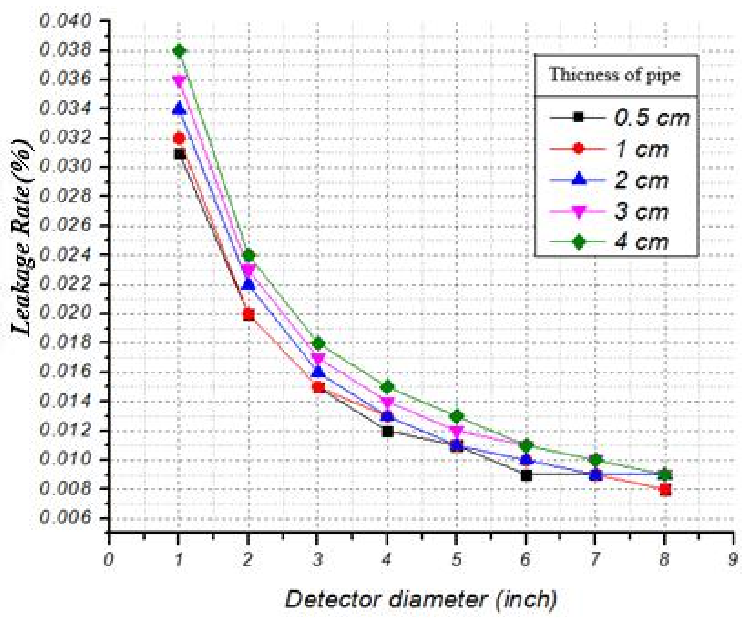

Figure 9. The effect of the polyethylene thickness (pipe diameter 4 inch) on the detectable leakage using ^{99m}Tc with activity of 10 mCi.

the detector. The effect of pipe diameter (thickness of 0.5 cm) has been investigated on the amount of leakage detection in figures 10 and 11.

For detectors with dimensions of 3×3 inch and pipe diameters of 2, 4, 6 and 8 inches, leakages more than 0.02%, 0.025%, 0.03% and 0.034% of inlet fluid volume for iron pipe and leakages more than 0.013%, 0.015%, 0.017% and 0.019% of inlet fluid volume for polyethylene pipe can be distinguished, respectively.

3.4. Effect of radiotracer activity in the leakage detection and determination of its amount

One of the most important parameters in leakage detection is the activity of radiotracer. The increase in activity causes NAUC to be more sensitive to less leakage, because for constant leakage, the difference between NAUC of two detectors can be distinguished. Figure 12 shows the effect of activity in the amount of detectable leakage for ^{99m}Tc.

For example, in iron pipe (outer diameter of 4 inches, wall thickness of 0.5 cm, detector of 3x3 inch, petrol as a fluid and ^{99m}Tc as a tracer), the detectable leakages is equal to 2.422% and 0.008% of inlet fluid volume can be determined by activity of 1 μCi and 100mCi, respectively. It means that using more activity leads more sensitivity in the leakage measurement.

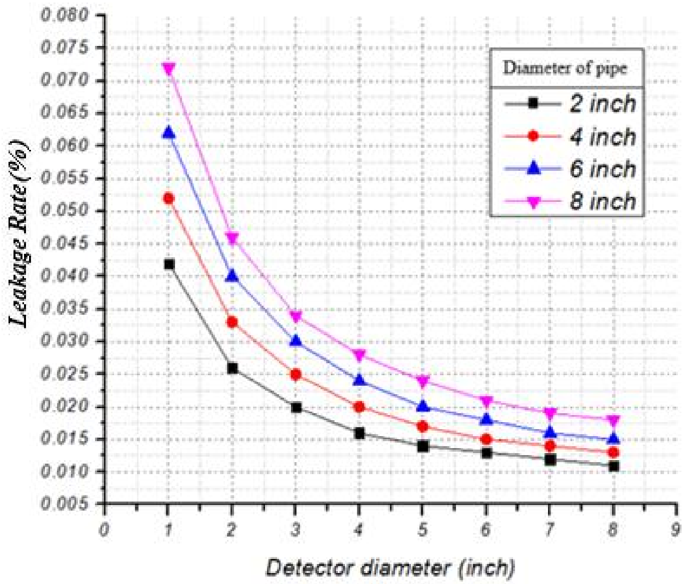

Figure 10. The effect of the iron diameter (pipe diameter 4 inch) on the detectable leakage using ^{99m}Tc with activity of 10 mCi.

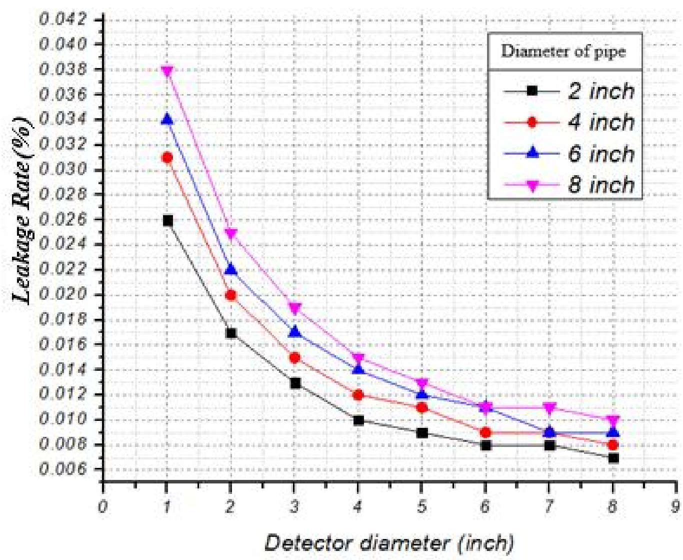

Figure 11. The effect of the polyethylene diameter (diameter pipe 4 inch) on the detectable leakage using ^{99m}Tc with activity of 10 mCi.

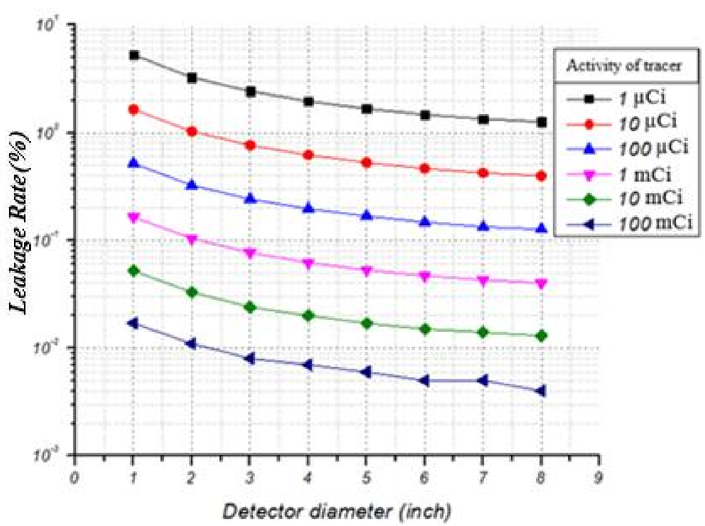

Figure 12. The effect of tracer activity on the detectable leakage using ^{99m}Tc in iron pipe (wall thickness of 0.5, diameter pipe of 4 inch).

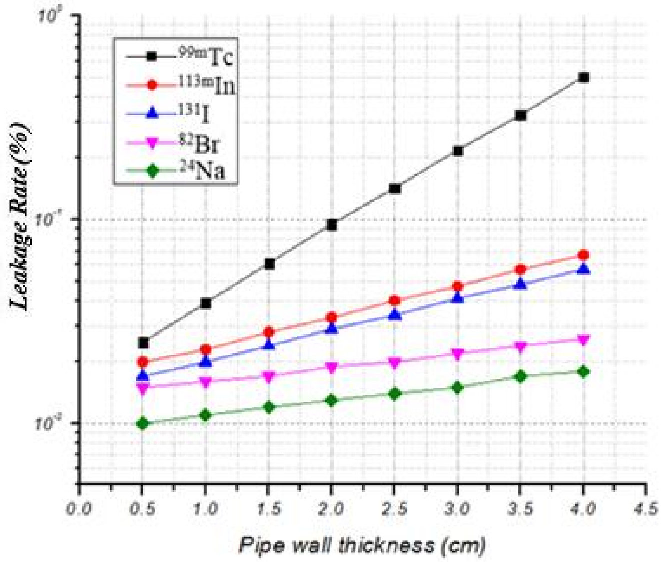

Figure 13. The effect of different tracers in the amount of leak detection in iron pipe with different thicknesses (diameter pipe of 4 inch).

3.5. Effect of different radioactive tracers in the leakage detection and determination of its amount

Various liquid radioactive tracers (^{24}Na, ^{82}Br, ^{131}I, ^{99m}Tc, ^{113m}In) have been used for our purpose (Table 1). Two important characteristics of radiotracer are energy of emitted gamma as well as ratio of gamma emission in decay scheme.

Accuracy of leakage detection is dependent on the number of photons reached to the detector and recorded by it. For high accuracy detection in pipes with high atomic number material (iron), radioactive tracers with high gamma energy should be used. In pipes with low atomic number, radiotracer with low energy gamma is more appropriate, because low energy photons can be recorded in detector with higher efficiency.

The amount of leakages has been measured with different radiotracers (iron pipe, outer diameter of 4 inches, fluid petrol, and activity of 10 mCi). As shown in figure 13, by increasing the wall thickness, the slope curve of the ^{99m}Tc is greater than of the other tracers, it means that using ^{99m}Tc, the capability of leakage measurement is reduced due to its low energy.

In addition, ^{131}I (with HVL and ratio of gamma emission equal to 0.909 cm and 89% (Table 1), respectively [10]) and ^{113m}In (with HVL and ratio of gamma emission equal to 0.963 cm and 65% (Table 1), respectively [10]) can measure equal amounts of leakage (larger HVL but less emitted gamma for ^{131}I) as shown the same slope. However, The slope curve of ^{82}Br, due to higher gamma emission as well as higher energy, is smaller than the slope curve of ^{113m}In and ^{131}I, so it will be able to measure more leakages.

By increasing the thickness of the iron pipe, small changes in slope curve of ^{24}Na will be caused, so it has the ability to measure less leakage (the best in leakage distinguishing). This is due to the emission of two high energy gammas with a ratio of gamma emission equal to 100% (Table 1).

On the other hand, curve slope reflects the sensitivity of the amount of leakage to the thickness of the pipe.

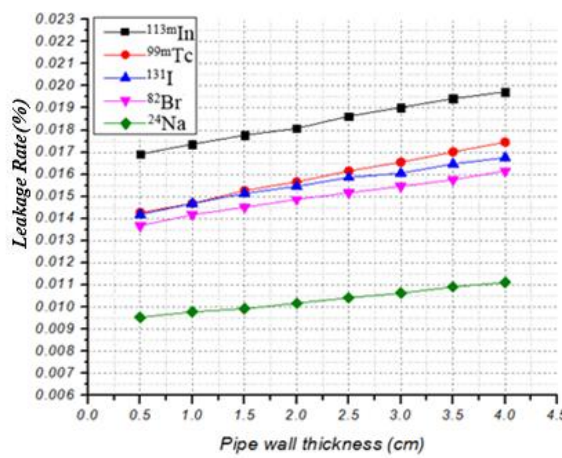

Figure 14. The effect of different tracers in the amount of leak detection in polyethylene pipe with different thicknesses (diameter pipe of 4 inch).

Smaller slope indicates better measurement independent from thickness. As a result, based on the fluid, type, thickness, diameter of the pipe as well as the required accuracy for leak detection, different radiotracers with different activities can be selected. For example, for determination of leakage in iron pipes (wall thickness of 1 cm, diameter of 4 inch, petrol as a fluid, using 3x3 inch detector and activity of 10 mCi) leakages more than 0.061%, 0.028%, 0.024%, 0.017% ,0.012% of inlet fluid volume can be distinguished by ^{99m}Tc, ^{113m}In ,^{131}I, ^{82}Br and ^{24}Na, respectively (Figure 13). Also, in polyethylene pipe, leakages more than 0.014%, 0.017%, 0.014%, 0.014%, 0.009%, of input fluid volume can be distinguished for tracers ^{99m}Tc, ^{113m}In ,^{131}I, ^{82}Br and ^{24}Na, respectively (Figure 14)

4. Conclusion

According to the requirements for leakage detection in industrial pipelines, the detection of leak and determination of its amount using radioactive tracer injection method was investigated in this paper. The net area under curves of recorded count for the detector array including two NaI (Tl) was used for measurements. Also, the effect of material and thickness of pipe, activity of tracer and its type (^{24}Na, ^{82}Br, ^{131}I, ^{99m}Tc, ^{113m}In) as well as types of fluid have been investigated on the detectable amount of leakage.

By using radiotracer injection method, the effect of environmental conditions can be neglected on leak detection. It was shown that by increasing the activity of the radiotracer, the measurement of less leakage will be possible.

Also, for different conditions, minimum detectable leakages were investigated (Figures 6-14). These results can be used as a tool for selecting the set up for leakage detection based on the specified conditions as well as the considered confidence level (based on the fluid, type, thickness, diameter of the pipe as well as the required accuracy).

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